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Oxidation of aluminum plates MTR fuel in thermal transfer conditions

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Abstract

The mechanism of aluminium oxidation has been studied in relation with the in-reactor behaviour or nuclear fuel plates in MTR reactors. Experiments have been carried out to compare the oxidation level with and without heat transfer. A double loop device was used to simulate the fuel behaviour, which heats an aluminium fuel channel from the outer sides while cooling the inside by circulating demineralised water. Also, a rotating aluminium cylinder was exposed in demineralised water at temperature in an autoclave. Both experiments have been performed at the same plate temperature, with the same water velocity with respect to the metal surface. The oxide produced in the experiment with heat transfer is significantly thicker than that obtained in the rotating autoclave, what indicates an important role of the thermal gradient in the oxidation mechanism.

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Keywords: Research Reactor; Aluminum Oxidation; Plate Type-Fuel; Rotating Autoclave; Thermal Gradient.

1. Introduction

According to data published by the International Atomic Energy Organization, OIEA (2014), there are 247 Research Nuclear Reactors (RR) operating in the world, six are being built and twelve more are being planned, the Argentine reactor project RA-10 among them. In many cases, a compacted mixture of particles of a uranium compound dispersed in an aluminum matrix is used as fuel. This fissile material is sheathed by co-lamination between two sheets of aluminum alloy (flat or slightly curved). The dimensions of the plates vary from one reactor

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to another but, approximately, are 600×60×1.5 mm, where the thickness of fissile compound does not exceed 500 microns. Groups of about twenty plates are parallel arranged and supported by two side brackets of inert material, to form what is called fuel element, which is finally installed in the reactor core at a given position.

During reactor operation, the generated heat is extracted by a high purity water flow through the channels formed between the fuel plates. Under these conditions aluminum alloys develop an oxide layer on all surfaces exposed to water, which -having low heat conductivity- hampers the heat transfer. This causes the increment of temperature of the metal-oxide interface and a feedback process is produced, in which the increase of interface temperature generates an acceleration of the oxidation of aluminum. In high flux reactors, the high linear power dissipated can generate temperatures in the aluminum-oxide interface high enough to cause intergranular corrosion, with blistering and peeling, this will cause the plates to lose their holding capacity, causing undesirable loss of nuclear material and fission products.

The oxide growth prediction is complex, requiring the knowledge of many variables: linear power, flow rate, inlet and outlet coolant temperature, conductivity and pH of water, radiation level, etc. The oxide growth predictions [Griess et al. (1964), Ondrejcin (1983), Pawel et al. (1995), Kim et al. (2008)] are only valid in the range of parameters in which they were developed. The latest published by Kim et al. (2008), although intended to be more flexible to contain more variables, happens to be very sensitive to variation of some parameters -especially pH-, which makes it not practical to predict a real situation in an open pool reactor (Fig. 1).

With the purpose of obtaining an improved prediction for the case of RA10 reactor and others of similar design, tests were performed to simulate nuclear fuel plates thermal and hydraulic conditions during in reactor operation, in order to measure the oxide thickness generated under controlled conditions of the variables which influence the phenomenon; noteworthy, one of the most important parameters of the oxidation process is water pH [Griess et al. (1964)]. In this work, tests have been performed without pH control, as happens in Argentine reactors, including RA-10. In these cases, due to carbonation, pH at 25 ºC normally fluctuates between 5.5 and 6.5 at the pool surface, although values of 7 are believed to exist in contact with the hot fuel plates [Haddad et al. (2012)]. Also, at the same time, this work aims to provide data to understand the mechanism of oxidation, concentrating on the influence of the thermal gradients generated by the heat transfer, through comparison with static tests.

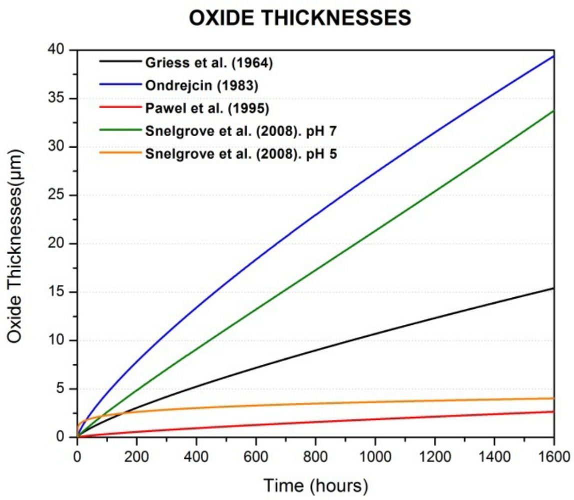

Fig. 1. Different aluminium oxide growth predictions, calculated for heat flux density of 2 MW/m^2, coolant speed of 8 m/s and plate temperature of 104 °C.

2. Experimental disposition

In order to study the effect of temperature gradient on the mechanism of oxidation, test comparing situations with and without heat transfer were mounted. For the results to be comparable, the test temperature in the second case was set at the first case's plate temperature (as in the thickness of the aluminum sheets) and so was done with the velocities of coolant water with respect to aluminum surfaces.

Tested alloy's composition is shown in Table 1; the material (AA6061 in the form of sheets) was provided by the RR Fuel Elements Manufacturing Plant (E.C.R.I.), located in the Constituyentes Atomic Center, Buenos Aires. Both the thermo-mechanical treatment and surface finishing reproduce those of RR fuel elements. The final treatment includes a pickling with sodium hydroxide at 70 °C, subsequent neutralization and rinsing.

Table 1. Composition of the aluminum alloy 6061.

Alloying	Mg	Si	Fe	Cu	Cr	Mn	Zn	Ti	Al
% Weight	0.95-1.10	0.55-1.10	0.45-0.15	0.2-0.4	0.1-0.2	0.1	0.25	0.03-0.07	Bce.

2.1. Tests without heat transfer

For tests without heat transfer a stainless steel rotary autoclave was used, which consist of a cylindrical container of 32cm high and 10cm in diameter, capable of working at temperature and pressure. It is warmed by a heating blanket and controlled by a Novus controller, model N480D-RPR220V. The device's main characteristic is that its top cover is crossed by a shaft which can rotate freely by means of a system of pulleys and belts connected to an electric motor at its upper end, while its lower end is equipped to fix the test piece; Fig 2. shows the device in operation.

The test piece was designed in order to have the tangential aluminum surface speed equal to the coolant speed in the heat transfer tests. Hence, its geometry takes into account the electrical motor rotating speed and the gear ratio, what yields a specimen diameter of 6 cm. Two aluminum disks of 5 mm thickness were used to assemble the cylinder, which were wrapped with a 10 × 19 cm aluminum plate which was given the corresponding cylindrical shape; Fig. 3.

Fig. 2. Operating rotary autoclave.

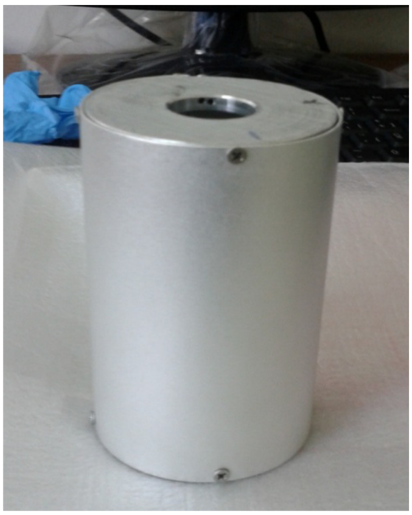

Fig. 3. Rotating aluminum cylinder.

2.2. Heat transfer device

To produce the required thermal gradient, a system that simulates the heat transfer that takes place in the nuclear fuels in operation was used. It is composed by an artificial channel of parallel plates of aluminum alloy through which the coolant water is made to flow at a specified speed. By the outside of the channel, the plates are in contact with a hot fluid, while inside circulates demineralized water, which acts as coolant as show in the scheme of Fig. 4(a) and (b).

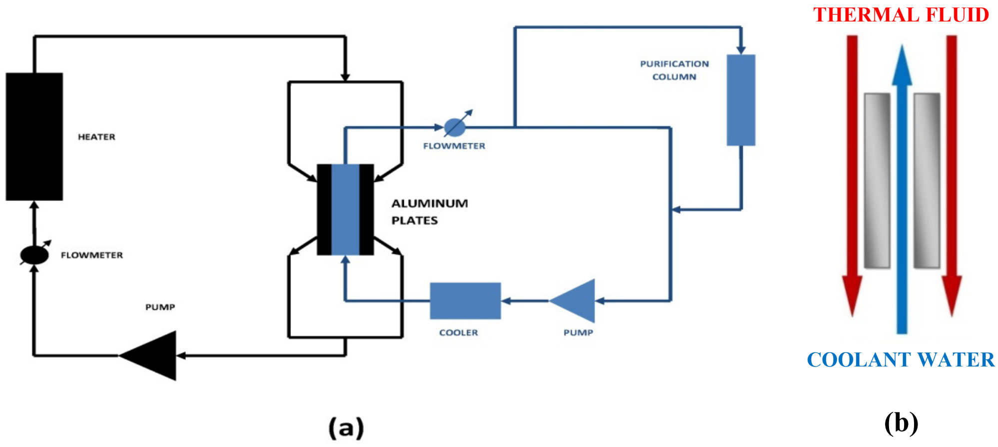

Fig. 4. (a) General scheme of the device; (b) Simulated channel.

The device is equipped with sensors to measure the most important process variables: temperature of thermal fluid, admission and discharge pressure of thermal fluid pump, inlet and outlet temperature of the coolant, coolant flow; internal plates temperature (by means of thermocouples inserted in the thickness) and other. The test channel measures 31 cm long, 4 cm wide and 2.5 mm thick. The heat is transferred in the middle of the channel, through a window of 10 cm long and 4 cm wide.

Fig. 5 shows the experimental device and Fig. 6 a detail of the fluid inputs and outputs and plates holder.

Fig. 5. Device overview

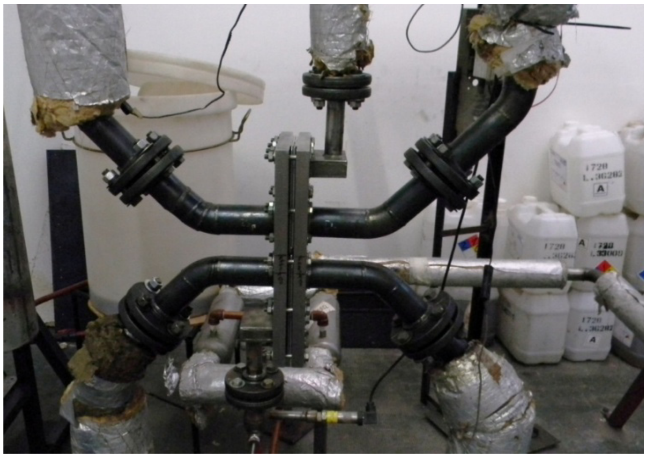

Fig. 6. Detail of plate holder

3. Results

Tests were performed with and without heat transfer with identical plate temperature and water flow, in order to make a comparison between the two cases. Then, oxide thickness was measured by the eddy current technique, at the Department of Non Destructive Testing (E.N.D.E.) of the Constituyentes Atomic Center, using a Fisher DualScope MP40E-S equipment. This technique was calibrated by Haddad et al. (2008) through comparison with scanning electron microscopy (SEM) and optical images.

3.1 Test with heat transfer

Three tests were performed lasting different periods of time, 96 hours the first, the second 600 hours and the third 1200 continuous hours. Experimental parameters were adjusted to regulate the initial plate temperature at 100 °C. During the course of the test, the plates suffer a heating due to oxide growth, which acts as a resistance to heat flow, causing an increase of aluminum plate temperature.

The most important conditions of the tests are listed below:

- Heat flux density: 4.67 MW/m^2
- Coolant velocity: 5.94 m/s
- Refrigerant inlet temperature to the channel: ~ 50 °C
- Temperature difference between inlet and outlet water: 4 °C
- Coolant conductivity: < 1 µS/cm
- Thermal fluid flow: 24.5 m^3/h

The plates were identified as Left Plate "I" and Right Plate "D", according to their position respect to the plate carrier. The oxide thickness was measured from the entrance zone to the exit, every 2 centimeters, except in the central zone (transfer window) where the interval was 1 centimeter, 6 measurements were performed per position, two in each side and two in the middle of the channel. Fig. 7 depicts the appearance of the three tested plates.

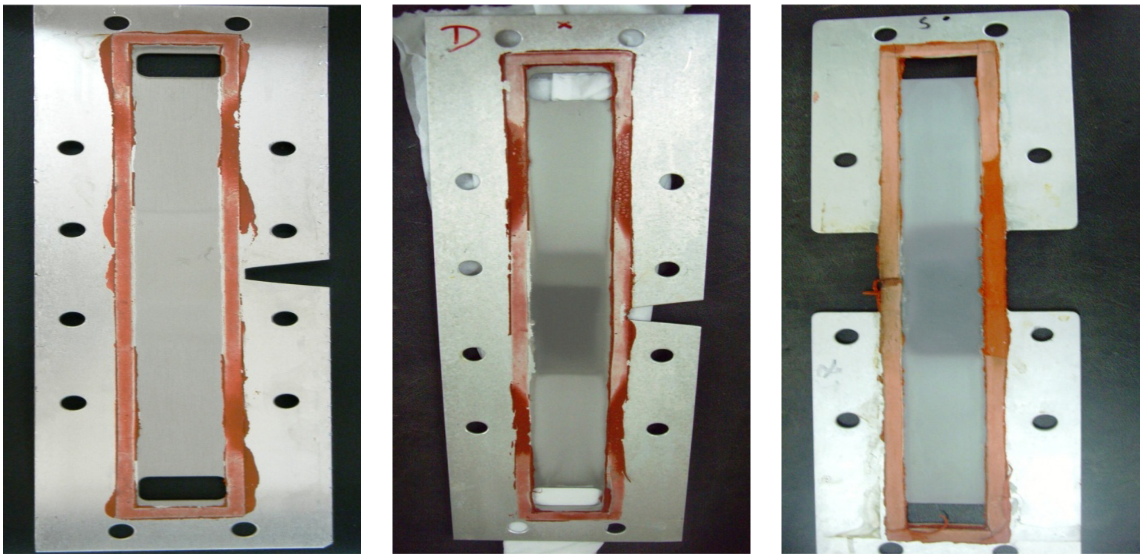

Fig. 7. (a) Plate tested at 96 hours; (b) Plate tested at 600 hours; (c) Plate tested at 1200 hours.

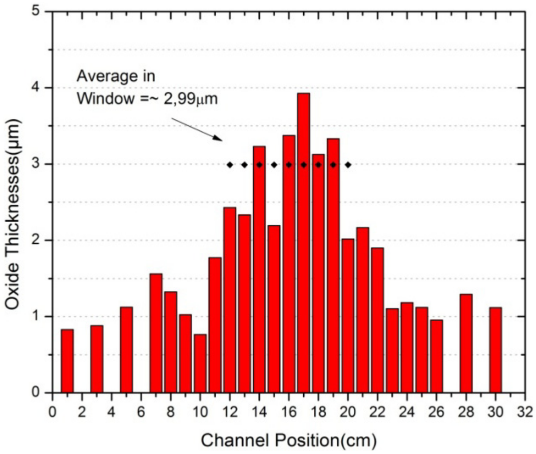

Fig. 8. Histogram of oxide thickness at 96 hours.

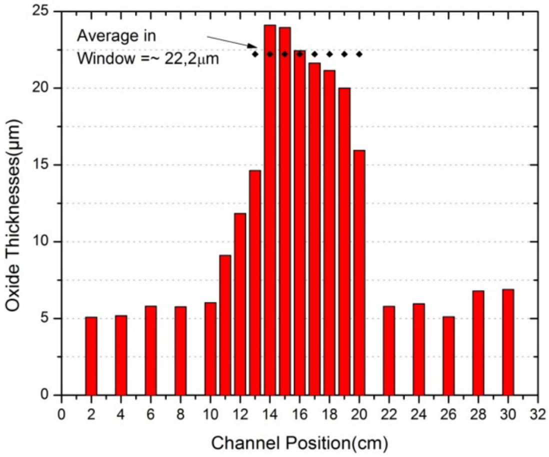

Fig. 9. Histogram of oxide thickness at 600 hours.

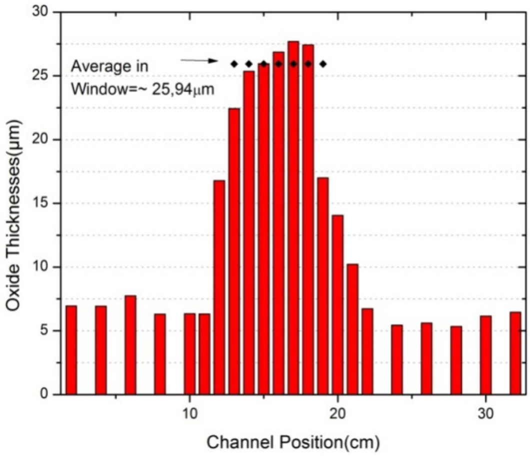

Fig. 10. Histogram of oxide thickness at 1200 hours.

In Figs. 8, 9 and 10 the histograms of oxide thickness measurements along the channels are detailed.

3.2 Test without heat transfer

Two tests were performed, the first with a duration of 96 hours and the second of 600 continuous hours; a future test of 1200 hours is being planned to complete the comparison. These tests were carried out in the rotary autoclave in demineralized water (conductivity < 1 μS/cm). Having a rotation speed of 1420 rpm and a cylindrical specimen with a radio of 3 cm, a tangential velocity of 4.45 m/s is reached in the cylinder surface. The autoclave temperature was set to reproduce the plate temperature of the test with heat transfer, including the daily increment due to the oxidation process. Fig. 11 shows the specimens aspect after the tests: (a) 96 hours and (b) 600 hours.

Fig. 11. (a) Cylindrical specimen tested for 96 hours; (b) cylindrical specimen tested for 600 hours.

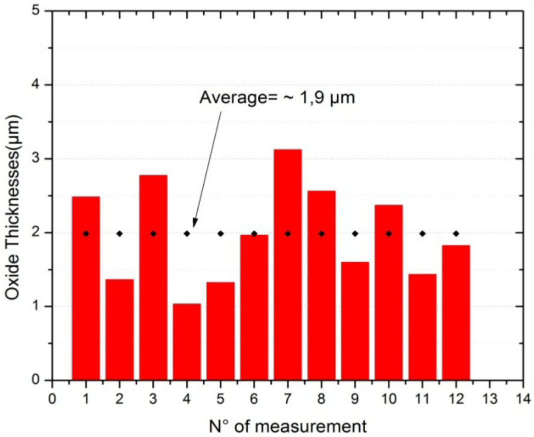

Fig. 12. Histogram of oxide thickness at 96 hours.

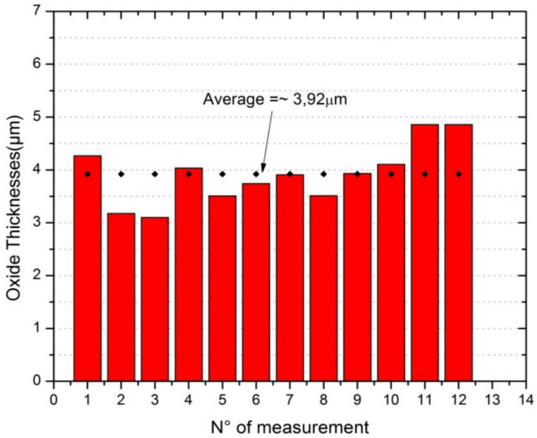

Fig. 13. Histogram of oxide thickness at 600 hours.

The thickness measurements were performed by the eddy current method in the central zone of the cylinder, in an area of approximately 6 × 6.5 cm. This area has a more regular oxide, because it is not affected by the flow disturbance. Twelve measurements were made in this area, repeating each measurement twice.

In Figs. 12 and 13 the detailed histograms of oxide thickness measurement in these areas are shown.

4. Discussion and Conclusions

The growth of aluminum oxide was compared after exposure to demineralized water with and without heat transfer, keeping the same aluminum plate temperature and velocity of water with respect to the metal surface (5 m/sec).

The growth of the aluminum oxide thickness in both conditions is shown in Fig. 14. As it can be seen, the values obtained under conditions of heat transfer are much greater than those generated in the system with uniform temperature.

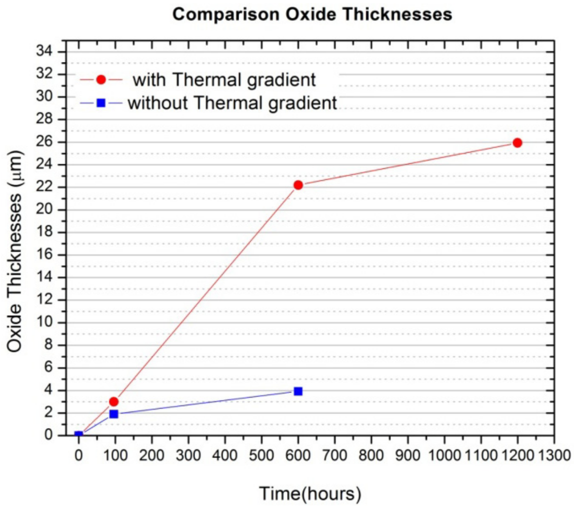

Fig. 14. Temporal evolution of oxide thickness tests with and without thermal gradient.

Being the water temperature in the autoclave higher than the coolant temperature in the channel in device with thermal gradient (100 °C against 50 °C), the viscous boundary layer is thinner, which favors the diffusional processes, such as oxidation, which depends on the migration of oxygen and/or metal ions. Therefore, it would be expected that the oxide growth be higher in the autoclave, which is not true. These considerations suggest that the presence of the thermal gradient generated during the heat transfer through the aluminum plate accelerate the growth of the oxide layer. As previously mentioned, the expected test at 1200 hours in autoclave will serve to corroborate this trend.

The experimental facility that generates the thermal gradient is able to reproduce the situation of the nuclear fuels in an experimental reactor in operation. It can register and control various parameters in real time and can work under operating conditions corresponding to reactors of different characteristics and different regimes. This feature makes the system a potentially useful tool to produce the necessary data to generate the correlations used to predict the behavior of the fuel within the reactor.

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